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(54) Title: BORON-LOW Nd-Fe-B ALLOY AND METHOD FOR PRODUCING PERMANENT MAGNETS ON THE BASIS OF SAID ALLOY

[see source for graphic]

(57) Abstract: The invention relates to boron-low Nd-Fe-B permanent magnets that have high coercive forces. To this end, the concentrations have to meet the following requirements:  $26.9 \text{ wt. \%} \leq [\text{Co}] \leq 5 \text{ wt. \%}$ ;  $0.05 \text{ wt. \%} \leq [\text{Co}] \leq 0.3 \text{ wt. \%}$ ;  $0.05 \text{ wt. \%} \leq [\text{Ga}] \leq 0.35 \text{ wt. \%}$ ;  $0.02 \text{ wt. \%} \leq [\text{Al}] \leq 0.3 \text{ wt. \%}$ .

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*For an explanation of the two-letter codes, and other abbreviations, please refer to the explanations ("Guidance Notes on Codes and Abbreviations") at the top of each regular issue of the PCT Gazette.*

### Description

Boron-low Nd-Fe-B alloy and method for producing permanent magnets on the basis of said alloy.

### Field of the Invention

The invention relates to an alloy on the basis of at least one rare earth, of at least one transition metal and boron, as well as to a method for producing permanent magnets on the basis of said alloy.

### Background of the Invention

Such alloys and methods for producing permanent magnets on the basis of this alloy are known from EP-A-0 124 655. In the known method, an alloy on the basis of neodymium, iron, and boron is initially melted. The alloy is then melted to a melting block, which is then reduced to powder. Blanks are pressed from this powder in a magnetic field, which are then sintered.

For many applications of Nd-Fe-B permanent magnets, especially in motors and all sorts of drives, the coercivity  $H_{CJ}$  of 150°C is key to the quality of the permanent magnet. At a low counter field load, the coercivity  $H_{CJ}$  at 150°C must be at least 4.5 kOe, better yet more than 5 kOe. At a high counter field load, values above 13 kOe at 150°C are even required. In addition to the high coercivity  $H_{CJ}$ , such magnets should also possess a remanence  $B_r$  that is as high as possible. For example, the remanence  $B_r$  of Nd-Fe-B permanent magnets that have a coercivity  $H_{CJ}$  within the area of 4.5 kOe, should be at least 1.29 T at ambient temperature, but better yet more than 1.35 T.

Applications in motors demand the additional requirement that the reversible temperature coefficient of the remanence  $TK (B_r)$  must be better than  $-0.11 \text{ \%}/\text{K}$  within a temperature range of  $20^\circ\text{C}$  to  $150^\circ\text{C}$ . Also, such permanent magnets should possess a corrosion

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resistance that is as good as possible, in order to eliminate complicated and expensive coatings. It is required, for instance, that the loss of mass of uncoated magnets in a so-called HAST test should be lower than  $1 \text{ mg}/\text{cm}^2$  after ten days. In the HAST test, permanent magnets are exposed to a pressure of 39.16 psi at an ambient temperature of  $130^\circ\text{C}$  and a relative humidity of 95%.

These requirements are currently not being met by conventional Nd-Fe-B permanent magnets.

#### Summary of the Invention

Based on this prior art, the invention is based on the task of creating an alloy for permanent magnets on the basis of at least one rare earth, and of at least one transition metal and boron that has a higher coercivity  $H_{CJ}$  than conventional alloys at the same remanence  $B_r$ , and possesses a lower temperature coefficient of the remanence, and is corrosion resistant.

~~This task is solved by the invention by the use of an alloy possessing the characteristics stated in claim 1.~~

Conventional Nd-Fe-B alloys essentially consist of three phases: the magnetically hard  $\phi$ -phase with the compound  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , the nonmagnetic  $\eta$ -phase with the compound  $\text{Nd}_{1.1}\text{Fe}_4\text{B}_4$ , and the nonmagnetic filler phase, which almost exclusively consists of Nd. The Nd-rich filler phase separates the particles of the  $\phi$ -phase from each other magnetically, which results in a high coercivity  $H_{\text{CJ}}$ . However, in a B concentration that is too low causes the risk of formation of a magnetically soft  $\text{Nd}_2\text{Fe}_{17}$  phase instead of the nonmagnetic  $\eta$ -phase, which substantially reduces the coercivity  $H_{\text{CJ}}$ . Unlike in conventional Nd-Fe-B alloys, the alloys produced according to the invention, the  $\text{Nd}_2\text{Fe}_{17}$  phase harmful for the coercivity  $H_{\text{CJ}}$  is not created instead of the nonmagnetic  $\eta$ -phase when a critical B-content is not reached, but instead initially a series of

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nonmagnetic phases containing Ga. Contrary to the ferromagnetic  $\text{Nd}_2\text{Fe}_{17}$  phase, these phases containing Ga contribute to the magnetic isolation of the particles of the  $\phi$ -phase, thus improving the coercivity  $H_{\text{CJ}}$  and also the temperature dependency of the alloy.

The invention is further based on the task of stating a method for producing permanent magnets on the basis of said alloy.

This task is solved by the invention by means of a method with the characteristics listed in claim 6.

Particularly high values can be achieved for the coercivity  $H_{CJ}$  with a skillfully adjusted temperature. It should be highlighted that particularly well values can be achieved for the coercivity  $H_{CJ}$  especially with rapid cooling. Rapid cooling, however, goes hand in hand with an effective use of the ovens. Therefore, even large permanent magnet components can be produced with slow cooling, without the formation of cooling tears in the permanent magnet components, and without a substantial decrease of the coercivity  $H_{CJ}$ .

#### Brief Description of the Drawings

The invention is explained in detail in the following attached drawings. They show:

- Figure 1      a section from a phase diagram for Nd-Fe-B permanent magnets;
- Figure 2      an illustration of the correlation between remanence  $B_r$  and coercivity  $H_{CJ}$  for various Nd-Fe-B permanent magnets;
- Figure 3      a diagram with a temperature adjustment during the sintering and starting annealing procedures;

- Figure 4 an additional diagram with an additional possible temperature adjustment during the sintering and ~~starting~~annealing procedures;
- Figure 5 an illustration showing the dependency of the coercivity  $H_{CJ}$  from the type of temperature adjustment during the sintering and ~~starting~~annealing procedures;
- Figure 6 a diagram showing the dependency of the remanence  $B_r$  from the effective contents of boron and rare earths;
- Figure 7 an illustration showing the dependency of the coercivity  $H_{CJ}$  at 150°C of the effective content of boron and rare earths during slow cooling;
- Figure 8 an illustration showing the dependency of the temperature coefficient of the coercivity TK ( $H_{CJ}$ ) from the effective content of boron and rare earths during slow cooling;
- Figure 9 an illustration showing the dependency of the coercivity  $H_{CJ}$  at 150°C of the effective content of boron and rare earths during rapid cooling; and

Figure 10 an illustration showing the dependency of the temperature coefficient TK ( $H_{CJ}$ ) of the coercivity  $H_{CJ}$  of the effective content of boron and rare earths with rapid cooling.

### Detailed Description

Figure 1 is a phase diagram showing the composition of an Nd-Fe-B alloy in dependency of the effective content of boron and rare earths. The structure suitable for the application as a permanent magnet particularly exists within a phase triangle 1. The alloy consists of magnetically hard particles of the  $\phi$ -phase with the composition  $Nd_2Fe_{14}B$  within this phase triangle 1, as well as of particles of the nonmagnetic  $\eta$ -phase with the composition

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$Nd_{1.1}Fe_4B_4$ , and the nonmagnetic filler phase, which almost exclusively consists of Nd. The Nd-rich filler phase separates the particles of the  $\phi$ -phase from each other magnetically, which is necessary in achieving a high coercivity  $H_{CJ}$ .

In order to evaluate whether a certain compound of the alloy exists inside or outside of the phase triangle 1, it is initially necessary to correct the content of rare earths and boron with regard to contaminations, as a part of the Nd is bound in the form of Nd-oxides, Nd-nickel-carbides, and Nd-nitrides. The effective content of rare earth  $[SE]_{eff}$ , and the effective content of boron  $[B]_{eff}$  is derived from the following formulas:



$$[\text{SE}]_{\text{eff}} = ([\text{SE}] - [\Delta \text{SE}]) f,$$

$$[\text{B}]_{\text{eff}} = [\text{B}] f,$$

whereby  $[\text{SE}]$  and  $[\text{B}]$  each are the weight parts of rare earth and boron.  $[\Delta \text{SE}]$  is therefore the part of rare earth that is bound in the compounds  $\text{Nd}_2\text{O}_3$ ,  $\text{Nd}_2\text{CO}$ , and  $\text{NdN}$ .  $f$  is a standardization factor.

$$[\Delta \text{SE}] = 5.993 [\text{O}] + 16.05 [\text{C}] + 10.30 [\text{N}]$$

$$f = 100 / ([100 - [\Delta \text{SE}] - [\text{O}] - [\text{C}] - [\text{N}]]).$$

$[\text{O}]$ ,  $[\text{C}]$ , and  $[\text{N}]$  are the weight parts of O, C, and N. All concentration values in the formulas above are in weight-%.

The effective content of rare earth and boron influences the composition of the structure. In point  $\eta$  of the phase triangle 1, the structure exists almost exclusively in the form of the  $\eta$ -phase. In point  $\phi$  of the phase triangle 1, the alloy is in the  $\phi$ -phase, while in point SE it essentially consists of the Nd-rich filler phase. Principally, the part of the  $\eta$ -phase can be of any low value. With a boron content that is too low, however, there is a risk that

The magnetically soft  $\text{Nd}_2\text{Fe}_{17}$ -phase forms instead of the nonmagnetic  $\eta$ -phase, which substantially reduces the coercivity  $H_{CJ}$ . The compound of the Nd-Fe-B permanent magnets has therefore traditionally always been chosen so that it lies within the phase triangle 1, especially above the tie-line 2. The values for each point in the phase diagram in figure 1 are listed in table 1 (with any commas in any of the tables representing decimal points).

	SE in wt. %	B content in wt. %	Fe content in wt. % residue
	SE in Gew.% <i>wt %</i>	<i>Content in wt %</i> B-Gehalt in Gew.%	<i>Content in wt %</i> Fe-Gehalt in Gew.%
$\phi$	26,68	1,000	<i>residue</i> Rest
$\eta$	37,3	10,2	<i>residue</i> Rest
Nd-reich <i>rich</i>	98	0	<i>residue</i> Rest
$\text{Nd}_2\text{Fe}_{17}$	23,3	0	<i>residue</i> Rest

Table 1

For many applications of Nd-Fe-B permanent magnets, however, and especially in motors and all sorts of drives, the coercivity  $H_{CJ}$  is essential. The coercivity  $H_{CJ}$  of the Nd-Fe-B permanent magnet used should be at least 4.5 kOe, better yet at least 5 kOe with low counter field load. Higher values above 13 kOe at 150°C are required with higher counter field loads. In addition to a high coercivity  $H_{CJ}$  at a temperature of 150°C, such Nd-Fe-B permanent magnets should also have a remanence  $B_r$  that is as high as possible.

Particularly for the application in motors it is required that the reversible temperature coefficient of the remanence  $TK(B_r)$  within the temperature range of 20°C to 150°C should be better than  $-0.11 \text{ \%}/K$ .

Additionally, Nd-Fe-B permanent magnets should have a corrosion resistance that is as well as possible, in order to eliminate complicated and expensive coating.

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It was discovered that a phase area 3 forms by adding gallium to the alloy below the tie-line 2, by the fact that in addition to the magnetically hard  $\phi$ -phase, and in addition to the nonmagnetic, Nd-rich phase, further phases containing Ga exist. A tie-line 4 separates the phase area 3 from another phase area 5, because the  $Nd_2Fe_{17}$ -phase is predominant. Surprisingly, it is now possible to meet the requirements of the use of Nd-Fe-B permanent magnets in motors with the alloys in the phase area 3. This improvement can be explained by the following metallurgic model: In conventional Nd-Fe-B permanent magnets, the magnetically soft  $Nd_2Fe_{17}$ -phase that is harmful for the coercivity  $H_{CJ}$ , if the critical boron content falls short, as illustrated by the border line 2. When adding gallium, cobalt, and copper to the Nd-Fe-B alloy, not the  $Nd_2Fe_{17}$ -phase is created instead of the nonmagnetic  $\eta$ -phase with the values falling short of the border line 2, but initially a series of nonmagnetic phases containing Ga are created. Contrary to the  $Nd_2Fe_{17}$ -phase, these phases containing Ga contribute to the magnetic isolation of the particles of the

$\phi$ -phase. This improves the coercivity  $H_{CJ}$ , and also its temperature coefficient. A further reduction of the boron contents finally leads to the formation of the  $Nd_2Fe_{17}$ -phase in the phase area 5, and therefore to the breakdown of the coercivity  $H_{CJ}$ .

In addition to gallium, Co and Cu may also be added to the alloy with beneficial effect.

For example, by adding Co to the alloy, the temperature coefficient of the remanence TK ( $B_r$ ) of Nd-Fe-B permanent magnets can be improved. In particular the temperature coefficient of the remanence TK ( $B_r$ ) can be improved from  $-0.12\text{ \%}/K$  to approximately  $-0.105\text{ \%}/K$  by adding 3 wt.-% of Co to the alloy. However, if only Co is added to the alloy, this leads to the formation of a magnetically soft  $SECo_2$ -Laves-phase, thereby

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drastically reducing the coercivity  $H_{CJ}$ . The formation of this harmful Laves-phase can be prevented by adding Cu at the same time. It has been found that the addition of 0.05 to 0.2 wt.-% of Cu is beneficial. Furthermore, Nd-Fe-B permanent magnets containing Cu can be slowly cooled after a heating treatment performed during the production process, without essentially reducing the coercivity  $H_{CJ}$ .

The resistance of Nd-Fe-B permanent magnets to corrosion by water vapor is improved by about three orders of magnitude by additionally adding Co, Cu, and Ga to the alloy as opposed to conventional Nd-Fe-B permanent magnets. In this process, a particularly

reactive Nd-rich filler phase is largely replaced by chemically more noble phases containing Co, Cu, and Ga.

These measures result in Nd-Fe B permanent magnets showing a loss of mass of  $< 1 \text{ mg/cm}^2$  based on the surface of the Nd-Fe-B permanent magnet after ten days in a so-called HAST test. In the so-called HAST test, the Nd-Fe-B permanent magnets are exposed to a pressure of 39.16 psi at a temperature of 130°C and a relative humidity of 95%.

It is also possible to increase the coercivity  $H_{CJ}$  by replacing one part of the Nd with Dy, Tb, or Ho, without essentially changing the content ratio of rare earth to the Fe and B content. As, contrary to Nd, the magnetic moment of Dy, Tb, and Ho is oriented anti-parallel to the magnetic moment of Fe, this inevitably leads to a reduction of the achievable remanence  $B_r$ . This means that the increase of the coercivity  $H_{CJ}$  is tied to the decrease of the remanence  $B_r$ .

This connection is illustrated in figure 2 and the respective table 2.

Alloy—

Legierung Alloy	SE, effektiv	B, effektiv	Dy	Co	Cu	Ga	H <sub>CJ</sub> (20°C, kOe)	H <sub>CJ</sub> (150°C, kOe)	B <sub>r</sub> (20°C, T)
A 1	29,2	0,98	3	-	-	-	17	3,5	1,33
A 2	29,5	0,98	4,6	-	-	-	21	5	1,28
A 3	29,6	0,98	6,5	-	-	-	26	8	1,22
A 4	29,7	0,98	8,6	-	-	-	31	11	1,16
B 1	29,3	0,94	3	3	0,15	0,23	18	5,3	1,35
B 2	29,5	0,94	5,5	3	0,15	0,23	23	8	1,28
B 3	30	0,93	9,5	3	0,15	0,23	31	13	1,18

*effective*

Table 2

The alloys A1 to A4 represent conventional alloys with compounds as listed in table 2.

Alloys B1 to B3 are alloys according to the invention. Figure 2 clearly shows that although the coercivity increases with an increased content of Dy, the remanence decreases.

Furthermore, figure 2 shows that those alloys to which Co, Cu and Ga were added, have a higher coercivity H<sub>CJ</sub> at the same amount of remanence B<sub>r</sub> in comparison to conventional alloys. The latter does not apply to ambient temperature, but instead especially to a temperature of 150°C.

Nd-Fe-B alloys with a Dy content of around 3 wt.-% have now been examined systematically. The results of these tests are listed in tables 3 and 4. It was discovered during the course of these tests that the magnetic characteristics of Nd-Fe-B permanent

magnets largely depend on the temperature adjustment during the course of the heat treatment of the manufacturing process.

Nd-Fe-B alloys are usually produced by initially melting the alloy together with the desired compounds, and then decanted to a melting block. The melting block is then

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reduced to powder and possibly mixed with other powders to correct the final compound. The finished powder is then arranged in a magnetic field and pressed parallel or vertical to the magnetic field direction, or also by using isostatic pressure toward the blank. The blanks are then subjected to a sintering process 6, as illustrated in figures 3 and 4. In the example of the temperature adjustment illustrated in figure 3, a heat treatment 7 is performed after the sintering process 6. The cooling process of the ~~starting~~annealing temperature can be slow, as in figure 3, or rapid, as in figure 4.

Figure 5 shows the dependency of the coercivity  $H_{CJ}$  in dependence of the effective boron content and the cooling speed  $\Delta T/\Delta t$ . Figure 5 proves that a high coercivity  $H_{CJ}$  at a high boron content may only be achieved in a narrow temperature window between 440 and 500°C. However, higher amounts of coercivity  $H_{CJ}$  may be achieved at a lower effective boron content may be achieved at a larger temperature window. Therefore, the coercivity  $H_{CJ}$  increases by almost 3 kOe at decreasing boron content. The coercivity  $H_{CJ}$  can be additionally increased by approximately 1 kOe by a rapid cooling process of below

750°C during the course of the sintering process, and by rapid cooling of the starting temperature.

The high amounts of coercivity  $H_{CJ}$ , which result despite of slow cooling at a low effective content of boron of 0.92 wt.-%, are of particular interest. This is beneficial especially when the Nd-Fe-B permanent magnets containing large sectional surfaces are to be produced, because only low cooling speeds  $\Delta T/\Delta t$  of  $< 10$  K/min are permissible for such components during the sintering process and the heat treatment in order to avoid cooling rips. These low cooling speeds, however, may lead to slight imperfections of the magnetic characteristics only. According to figure 5 it is quite possible to slowly cool

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down Nd-Fe-B permanent magnets at cooling speeds within a range of 1 to 2 K/min after the heat treatment, without sustaining substantial imperfections of the magnetic characteristics, as long as the Nd-Fe-B alloy is low in boron. An Nd-Fe-B alloy low in boron is an alloy with an effective boron content of below the tie-line 2.

Tables 3 and 4 contain a list of compounds and magnetic characteristics of isostatically pressed Nd-Fe-B permanent magnets with various effective contents of rare earths and boron. The statements in bold font refer to the alloys low in boron content according to the invention. All Nd-Fe-B permanent magnets have been produced according to the common powder-metallurgic process, and were sintered to a density of  $> 7.6$  g/cm<sup>3</sup> at a



temperature of approximately 1060°C. The Nd-Fe-B permanent magnets listed in table 3 were slowly cooled to ambient temperature at approximately 1 to 2 K/min. They were then tempered at a temperature of 440°C to 560°C for one to two hours, and again slowly cooled to ambient temperature at approximately 1 to 2 K/min. The magnets listed in table 4 were initially quenched slowly from the sintering temperature to approximately 750°C at about 2 K/min, and after a waiting period of about 1 hour, they were rapidly quenched to ambient temperature at approximately 30 to 50 K/min. These Nd-Fe-B permanent magnets were then again rapidly cooled to ambient temperature at approximately 30 to 50 K/min after subsequent tempering at 470 to 530°C.

Figure 6 contains the values for the remanence  $B_r$  for the alloys in table 3 in dependency of the effective content of boron and rare earths. Two level lines illustrate the tendency of the increasing remanence  $B_r$  at decreasing effective rare earth content, and increasing effective boron content. A remanence  $B_r$  of over 1.35 T is achieved for isostatically pressed Nd-Fe-B permanent magnets with an effective rare earth content of < 30 wt.-%, and an effective boron content of > 0.93 wt.-%. With regard to the boron contents, the

remanence  $B_r$  goes through a maximum just below the threshold 2 to the phase triangle 1.

Figure 7 illustrates the dependency of the coercivity at 150°C for the slowly cooled Nd-Fe-B permanent magnets in table 3. Figure 7 proves that the coercivity  $H_{CJ}$  increases at 150°C with a decreasing effective boron content. The same applies for the coercivity at 20°C.

Finally, figure 8 shows the dependency of the temperature coefficient of  $H_{CJ}$  for slowly cooled Nd-Fe-B permanent magnets in dependency of an effective content of rare earths and boron. Here too, increasingly better values are achieved for the temperature coefficient with decreasing effective boron content. Together with the increasing coercivity  $H_{CJ}$ , this leads to an increase of the coercivity  $H_{CJ}$  at 150°C of below 4.5 kOe to values of up to above 5.5 kOe for slowly cooled magnets. These particularly high values for the coercivity  $H_{CJ}$  are especially beneficially particularly for a rare earth content  $[SE]_{eff}$  of more than 28.9 wt.-%, while the following relation applies for the effective boron content:

$$1.814 - 0.0303 [SE]_{eff} \leq [B]_{eff} \leq 1.396 - 0.01491 [SE]_{eff}$$

The same applies to Nd-Fe-B permanent magnets that are rapidly cooled from approximately 750°C and from ~~starting~~annealing temperature. However, according to

figures 9 and 10, slightly better values are achieved not only for the temperature dependency, but also for the absolute values as compares to the slowly cooled Nd-Fe-B permanent magnets. This results in an expanded area in which the required characteristics, namely a remanence  $B_r > 1.35$  T at ambient temperature, and a coercivity  $H_{CJ} > 5$  kOe at 150°C are achieved.

Particularly high values for the coercivity  $H_{CJ}$  at 150°C are achieved for an effective content of rare earth of above 28.5 wt./%, especially 28.7 wt./%, while the following

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relation applies to the effective boron content:

$$1.814 - 0.0303 [\text{SE}]_{\text{eff}} \leq [\text{B}]_{\text{eff}} \leq 1.478 - 0.01801 [\text{SE}]_{\text{eff}}$$

~~In conclusion it~~ It should be noted that Pr may also be used in addition to Nd, without negatively influencing the magnetic characteristics of the permanent magnets.

An alloy consistent with the present invention has at least one rare earth element (including yttrium), iron, B, Co, Cu, Ga, Al, and production-based contaminations, whereby the following relations apply to the effective rare earth content  $[\text{SE}]_{\text{eff}}$ , the effective boron content  $[\text{B}]_{\text{eff}}$ , the mutual content of Dy, Tb, and Ho  $[\text{Dy} + \text{Tb} + \text{Ho}]$ , the

cobalt content [Co], the copper content [Cu], the gallium content [Ga], and the aluminum content [Al]:

$$\underline{26.9 \text{ wt.}\% \leq [\text{SE}]_{\text{eff}} \leq 33 \text{ wt.}\%}$$

$$\underline{2.185 - 0.0442 [\text{SE}]_{\text{eff}} \leq [\text{B}]_{\text{eff}} \leq 1.363 - 0.0136 [\text{SE}]_{\text{eff}}}$$

$$\underline{[\text{Dy} + \text{Tb} + \text{Ho}] \leq 17 \text{ wt.}\%}$$

$$\underline{0.5 \text{ wt.}\% \leq [\text{Co}] \leq 5 \text{ wt.}\%}$$

$$\underline{0.05 \text{ wt.}\% \leq [\text{Cu}] \leq 0.3 \text{ wt.}\%}$$

$$\underline{0.05 \text{ wt.}\% \leq [\text{Ga}] \leq 0.35 \text{ wt.}\%}$$

$$\underline{0.02 \text{ wt.}\% \leq [\text{Al}] \leq 0.3 \text{ wt.}\%}$$

More particularly, the effective boron content may be defined by:

$$\underline{1.814 - 0.0303 [\text{SE}]_{\text{eff}} \leq [\text{B}]_{\text{eff}} \leq 1.363 - 0.0136 [\text{SE}]_{\text{eff}}}$$

or, if the rare earth content exceeds about 28.5wt%, by:

$$\underline{1.814 - 0.0303 [\text{SE}]_{\text{eff}} \leq [\text{B}]_{\text{eff}} \leq 1.478 - 0.01801 [\text{SE}]_{\text{eff}}}$$

Permanent magnets may be produced from alloys by performing the following:

- orienting in a magnetic field and pressing of powder into a blank;

- sintering of the blank at temperatures between about 1020°C and about 1140°C;

and

- cooling of the sintered blank to temperatures below about 300°C, whereby

cooling occurs at above about 800°C at a median cooling speed  $\Delta T_1/\Delta t_1$  of  $< 5 \text{ K/min}$ ;

and

whereby the following relation applies to the annealing temperature  $T_A$  of the sintered

blank in dependency of a median cooling speed  $\Delta T_2/\Delta t_2$ :

for  $\Delta T_2/\Delta t_2 < 5$  K/min:

$$450^{\circ}\text{C} \leq T_A \leq 550^{\circ}\text{C} \text{ for } [B]_{\text{eff}} < 2.993 - 0.069 [SE]_{\text{eff}}$$

$$460^{\circ}\text{C} \leq T_A \leq 510^{\circ}\text{C} \text{ for } [B]_{\text{eff}} > 2.993 - 0.069 [SE]_{\text{eff}}$$

for  $5 \text{ K/min} \leq \Delta T_2/\Delta t_2 \leq 100 \text{ K/min}$ :

$$450^{\circ}\text{C} \leq T_A \leq 550^{\circ}\text{C}.$$

After the sintering process, the blank may be maintained at a holding temperature between about 700 and about 800°C for a period of between about half an hour and 2 hours. Further, the blank may be cooled at a median cooling speed  $\Delta T_3/\Delta t_3$  of either  $> 5$  K/min or  $< 5$  K/min after the sintering process and maintenance at a holding temperature. Cooling speeds  $\Delta T_2/\Delta t_2$  and  $\Delta T_3/\Delta t_3$  may be between about 30-50 K/min, while cooling speeds  $\Delta T_1/\Delta t_1$  to  $\Delta T_3/\Delta t_3$  may be between about 1-2 K/min.

Compound in wt. %

starting

temp.

annealing

*effective*

Zusammensetzung in Gew. % Compound in wt %						Anneal- temp. (°C)	B <sub>r</sub> (20°C) (T)	(BH) max (MGOe)	H <sub>cJ</sub> (20°C) (kOe)	H <sub>cJ</sub> (150°C) (kOe)	TK(H <sub>cJ</sub> ) (20-150°C) (%/K)
SE, effektiv	B, effektiv	Dy	Co	Cu	Ga	(°C)	(T)	(MGOe)	(kOe)	(kOe)	(%/K)
28,1	0,99	2,8	3,1	0,15	0,22	470	1,386	46,6	16,16	4	-0,579
28,1	0,99	2,8	3,1	0,15	0,22	500	1,372	45,7	15,06		-0,570
28,1	0,99	2,8	3,1	0,15	0,22	530	1,382	46,4	15,57		
28,9	0,98	2,8	3	0,13	0,2	470	1,383	46,4	16,88	4,08	-0,583
28,9	0,98	2,8	3	0,13	0,2	500	1,378	46,1	17,24	4,39	-0,573
28,9	0,98	2,8	3	0,13	0,2	530	1,391	47,0	16,4	3,84	-0,589
29,6	0,97	2,8	2,9	0,1	0,18	470	1,376	46,0	16,27	4,02	-0,579
29,6	0,97	2,8	2,9	0,1	0,18	500	1,36	44,9	16,63	4,11	-0,579
29,6	0,97	2,8	2,9	0,1	0,18	530	1,374	45,8	9,96		
28,7	0,94	2,9	3,1	0,17	0,22	500	1,374	45,8	15,69	4,42	-0,553
28,65	0,95	2,9	3,1	0,16	0,22	500	1,356	44,6	16,43	4,51	-0,558
28,6	0,96	3	3,2	0,16	0,22	500	1,375	45,9	16,89	4,59	-0,560
28,55	0,97	3	3,2	0,15	0,22	500	1,375	45,9	17,58	4,42	-0,576
28,5	0,98	3	3,2	0,15	0,21	500	1,382	46,4	17,15	4,42	-0,571
29,8	0,92	3,1	3,0	0,16	0,22	500	1,341	43,6	18,08	5,36	-0,541
29,8	0,93	3,1	3,0	0,15	0,22	500	1,352	44,4	18,24	5,26	-0,547
29,8	0,95	3,1	3,0	0,15	0,22	500	1,355	44,6	18,11	5,04	-0,555
29,8	0,96	3,1	3,0	0,14	0,22	500	1,363	45,1	17,34	4,49	-0,570
29,8	0,98	3,1	3,0	0,14	0,22	500	1,348	44,1	17,42	4,41	-0,574
29,9	0,94	3,3	3,1	0,14	0,21	440	1,369	45,5	15,95	3,79	-0,586
29,9	0,94	3,3	3,1	0,14	0,21	470	1,342	43,7	17,71	4,67	-0,566
29,9	0,94	3,3	3,1	0,14	0,21	500	1,353	44,4	17,79	4,6	-0,570
29,9	0,94	3,3	3,1	0,14	0,21	530	1,352	44,4	10,62		
29,9	0,94	3,3	3,1	0,14	0,21	560	1,311	41,7	9,55		
29,2	0,93	2,9	3	0,19	0,25	470	1,364	45,2	16,56	4,67	-0,552
29,2	0,93	2,9	3	0,19	0,25	500	1,351	44,3	17	4,95	-0,545
29,2	0,93	2,9	3	0,19	0,25	530	1,366	45,3	16,38	5	-0,534
29,5	0,93	2,9	3	0,17	0,23	470	1,347	44,0	17,57	5,12	-0,545
29,5	0,93	2,9	3	0,17	0,23	500	1,331	43,0	18,21	5,39	-0,542
29,5	0,93	2,9	3	0,17	0,23	530	1,344	43,8	17,97	5,42	-0,537
29,9	0,92	2,9	3	0,16	0,22	470	1,341	43,6	18,62	5,42	-0,545
29,9	0,92	2,9	3	0,16	0,22	500	1,331	43,0	19,08	5,71	-0,539
29,9	0,92	2,9	3	0,16	0,22	530	1,307	41,5	18,56	5,61	-0,537

Table 3

Compound in wt. % \_\_\_\_\_ starting  
 \_\_\_\_\_ temp.

*annealing*

*effective*

Zusammensetzung in Gew.-% Compound in wt. %						Anlaß- temp.	B <sub>r</sub> (20°C)	(BH) <sub>max</sub>	H <sub>cJ</sub> (20°C)	H <sub>cJ</sub> (150°C)	TK(H <sub>cJ</sub> ) (20-150°C)
SE, effektiv	B, effektiv	Dy	Co	Cu	Ga	(°C)	(T)	(MGOe)	(kOe)	(kOe)	(%/K)
28,7	0,94	2,9	3,1	0,17	0,22	500	1,37	45,6	17,16	4,99	-0,546
28,65	0,95	2,9	3,1	0,16	0,22	500	1,341	43,6	18,02	5,15	-0,549
28,6	0,96	3	3,2	0,16	0,22	500	1,374	45,8	17,43	4,9	-0,553
28,55	0,97	3	3,2	0,15	0,22	500	1,372	45,7	16,33	4,61	-0,552
28,5	0,98	3	3,2	0,15	0,21	500	1,362	45,0	16,69	4,79	-0,551
29,8	0,92	3,1	3	0,16	0,22	500	1,343	43,8	18,3	5,59	-0,534
29,8	0,93	3,1	3	0,15	0,22	500	1,351	44,3	18,46	5,5	-0,539
29,8	0,95	3,1	3	0,15	0,22	500	1,35	44,2	18,17	5,18	-0,550
29,8	0,96	3,1	3	0,14	0,22	500	1,354	44,5	16,87	4,71	-0,554
29,8	0,98	3,1	3	0,14	0,22	500	1,344	43,8	16,91	4,78	-0,552
28,8	0,95	3	2,8	0,14	0,26	500	1,359	44,8	18,65	5,66	-0,536
28,8	0,95	3	2,8	0,14	0,26	530	1,361	45,0	18,22	5,67	-0,530
29,2	0,93	2,9	3	0,19	0,25	470	1,354	44,5	18,61	5,65	-0,536
29,2	0,93	2,9	3	0,19	0,25	500	1,343	43,8	18,87	5,67	-0,538
29,2	0,93	2,9	3,0	0,19	0,25	530	1,355	44,6	18,73	5,82	-0,530
29,5	0,93	2,9	3,0	0,17	0,23	470	1,342	43,7	19,71	5,83	-0,542
29,5	0,93	2,9	3,0	0,17	0,23	500	1,323	42,5	19,56	5,92	-0,536
29,5	0,93	2,9	3,0	0,17	0,23	530	1,329	42,9	19,9	6,09	-0,534
29,9	0,92	2,9	3	0,16	0,22	470	1,337	43,4	20,3	6,09	-0,538
29,9	0,92	2,9	3	0,16	0,22	500	1,343	43,8	19,8	5,9	-0,539
29,9	0,92	2,9	3	0,16	0,22	530	1,335	43,3	20	6,09	-0,535

Table 4

## Patent Claims

1. Alloy consisting of at least one rare earth, including yttrium, made of iron, consisting of the elements B, Co, Cu, Ga, and Al, as well as of production-based contaminations, whereby the following relations apply to the effective rare earth content  $[SE]_{eff}$ , the effective boron content  $[B]_{eff}$ , the mutual content of Dy, Tb, and Ho  $[Dy + Tb + Ho]$ , the cobalt content  $[Co]$ , the copper content  $[Cu]$ , and the gallium content  $[Ga]$ , as well as the aluminum content  $[Al]$ :

$$\begin{aligned} 26.9 \text{ wt. \%} &\leq [SE]_{eff} \leq 33 \text{ wt. \%} \\ 2.185 - 0.0442 [SE]_{eff} &\leq [B]_{eff} \leq 1.363 - 0.0136 [SE]_{eff} \\ [Dy + Tb + Ho] &\leq 17 \text{ wt. \%} \\ 0.5 \text{ wt. \%} &\leq [Co] \leq 5 \text{ wt. \%} \\ 0.05 \text{ wt. \%} &\leq [Cu] \leq 0.3 \text{ wt. \%} \\ 0.05 \text{ wt. \%} &\leq [Ga] \leq 0.35 \text{ wt. \%} \\ 0.02 \text{ wt. \%} &\leq [Al] \leq 0.3 \text{ wt. \%} \end{aligned}$$

2. Alloy according to claim 1,  
characterized in that  
the following relation applies to the effective boron content  $[B]_{eff}$ :

$$1.814 - 0.0303 [SE]_{eff} \leq [B]_{eff} \leq 1.363 - 0.0136 [SE]_{eff}$$

3. Alloy according to claims 1 or 2,  
characterized in that  
the rare earth content  $[SE]_{eff}$  is above 28.9 wt. %, whereby the following relation applies to the effective boron content:

$$1.814 - 0.0303 [SE]_{eff} \leq [B]_{eff} \leq 1.396 - 0.01491 [SE]_{eff}$$

4. Alloy according to claims 1 or 2,  
characterized in that  
the rare earth content  $[SE]_{eff}$  is above 28.5 wt. %, whereby the following relation applies to the effective boron content:



$$1.814 - 0.0303 [\text{SE}]_{\text{eff}} \leq [\text{B}]_{\text{eff}} \leq 1.478 - 0.01801 [\text{SE}]_{\text{eff}}$$

5. Alloy according to claim 4,  
**characterized in that**  
 the rare earth content  $[\text{SE}]_{\text{eff}}$  is above 28.7 wt. %.

6. Alloy according to one of the claims 1 to 5,  
**characterized in that**  
 the alloy has a Co content of between 2.5 and 3.5 wt. %.

7. Alloy according to one of the claims 1 to 6,  
**characterized in that**  
 the Cu content is between 0.1 and 0.2 wt. %.

8. Alloy according to one of the claims 1 to 7,  
**characterized in that**  
 the Ga content is between 0.20 and 0.30 wt. %.

9. Alloy according to one of the claims 1 to 8,  
**characterized in that**  
 the rare earths are selected from a group of the elements Nd, Pr, Dy, and Tb.

10. Method for producing permanent magnets from an alloy according to one of the claims 1 to 9 with the following process steps:  
 -orientation in the magnetic field and pressing of powder that was produced by reduction — of at least one melting body, into a blank;  
 -sintering of the blank at temperatures between 1020°C and 1140°C;  
 -cooling of the blank to temperatures below 300°C, whereby cooling occurs at above 800°C at a median cooling speed  $\Delta T_t / \Delta t_t$  of  $< 5 \text{ K/min}$ ; and

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—starting and cooling of the blank, whereby the following relation applies to the starting temperature  $T_A$  in dependency of a median cooling speed  $\Delta T_2/\Delta t_2$ :

for  $\Delta T_2/\Delta t_2 < 5$  K/min:

$$450^\circ\text{C} \leq T_A \leq 550^\circ\text{C} \text{ for } [B]_{\text{eff}} < 2.993 - 0.069 [SE]_{\text{eff}}$$

$$-460^\circ\text{C} \leq T_A \leq 510^\circ\text{C} \text{ for } [B]_{\text{eff}} < 2.993 - 0.069 [SE]_{\text{eff}}$$

—for  $5 \text{ K/min} \leq \Delta T_2/\Delta t_2 \leq 100 \text{ K/min}$ :

$$-450^\circ\text{C} \leq T_A \leq 550^\circ\text{C}.$$

11. Method according to claim 10,  
characterized in that

after the sintering process, the blank is maintained at a holding temperature between  $700$  and  $800^\circ\text{C}$  for a period of between half an hour and 2 hours.

12. Method according to claim 11,  
characterized in that

the raw body is cooled at a median cooling speed  $\Delta T_3/\Delta t_3$  of  $> 5$  K/min after the sintering process at holding temperature.

13. Method according to claim 12,  
characterized in that

the cooling speeds  $\Delta T_2/\Delta t_2$  and  $\Delta T_3/\Delta t_3$  are between 30 and 50 K/min.

14. Method according to claim 10 or 11,  
characterized in that

the raw body is cooled at a median cooling speed  $\Delta T_2/\Delta t_2$  of  $< 5$  K/min after the sintering process at holding temperature.

~~15. Method according to claim 14,~~  
**characterized in that**  
the cooling speeds  $\Delta T_1/\Delta t_1$  to  $\Delta T_2/\Delta t_2$  are between 1 and 2 K/min.

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[see source for fig. 1]

[Gew.% = wt.%]

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{see source for fig. 2}

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[see source for fig. 4 & 4]

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[see source for fig. 5]

[Gew.% = wt. %]

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[see source for fig. 6]

[Gew.% = wt.%]



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[see source for fig. 7]

[Gew. % = wt. %]

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[see source for fig. 8]

[Gew.% = wt.%]

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[see source for fig. 9]

[Gew.% = wt.%]

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[see source for fig. 10]

[Gew.% = wt. %]